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THE VISCOSITY OF LIQUID NITROGEN, CARBON MONOXIDE, ARGON AND OXYGEN AS A FUNCTION OF TEMPERATURE

N. S. Rudenko and L. W. Schubnikow
Ukrainian Physical-Technical Institute, Charkow, Ukraine, U. S. S. R.

ABSTRACT: The viscosity of liquid nitrogen, carbon monoxide, argon and oxygen as a function of temperature are measured in a temperature interval between the boiling point and the freezing point.

Experiments with condensed gases are almost completely absent among the /470* innumerable measurements of the viscosity of various liquids. The first measurement of the viscosity of liquid air at the boiling point was done by Forsch [1]; it resulted in a value of 0.0033 CGS units. Verschaffelt and Nicaise [2] measured the viscosity of impure oxygen and nitrogen as well as hydrogen and helium. For oxygen, nitrogen and mixtures of both, the following values were obtained:

TABLE 1.

Concentration (%)			Viscosity (CGS units)
N_2	O_2	Ar	
89	9.5	1.5	0.001599
1	96	3	0.001895
36	61	3	0.001865

The results of the two measurements apparently differ by a factor of 2.

We have undertaken the viscosity measurements in the course of a large-scale study of the properties of condensed gases and mixtures of gases since this information is important in the present development of cryogenic engineering. /471 We have investigated liquid nitrogen, carbon monoxide, argon and oxygen in the temperature interval between the boiling and the freezing point. At present we are investigating the viscosity of liquid methane and ethylene. We have decided to publish our results since they might be relevant for a future theory of viscosity.

Experimental Procedure

We have chosen to use the standard viscosimeter method in our measurements of the viscosity. This method has the advantage that the least amount of

*Numbers in the margin indicate pagination in the foreign text.

pure gas is required; large-scale production of pure gas would cause considerable difficulties. In addition, this method yields absolute values for the viscosity, and does not require knowledge of other constants, e.g. the density, which are not always known.

The viscosimeter (type "Ubellohde") was set up in a very long cylindrical Dewar vessel that was completely sealed by a cap (see Fig. 1). In order to produce the required temperatures, the vapors above the cooling agents (commercial oxygen and nitrogen) were evacuated and the pressure was regulated with a valve and vacuum gauge. The vessel was immersed in a second Dewar vessel which was filled with liquid air; thus evaporation was reduced and a constant temperature was maintained. Both vessels had a silver coating except for some narrow observation slits. The temperature was measured with a vapor-pressure thermometer that was mounted in close proximity to the viscosimeter (see Fig. 1). The liquid in the Dewar vessel was not agitated; however, after a few hours the temperature became constant everywhere in the vessel due to the extremely small flow of heat from the outside. At most, a small temperature difference could occur during rapid evacuation, but within the accuracy of the experiment this had no effect. The liquid in the viscosimeter was subjected to a positive pressure of approximately 150 mm Hg of an inert gas, thus preventing the liquid from boiling. Pure hydrogen was used as the inert gas since its solubility in the test gases is negligible and therefore could hardly influence the results. Both ends of the viscosimeter were connected to two bulbs via a system of three-way stop cocks. One bulb contained hydrogen of 50 - 150 mm Hg; the other contained hydrogen of 200 - 350 mm Hg. The pressure was indicated on a vernier scale with an accuracy of $\pm$ 0.1 mm. The discharge time of the liquid was between 6 and 30 min.; it was timed with a stop watch (accuracy: 0.2 sec). Most viscosity measurements were done with varying differences between the hydrogen pressures of the left and the right side of the viscosimeter. The results were always independent of the pressure. Each data point corresponds to the average of a series of between 3 and 5 measurements.

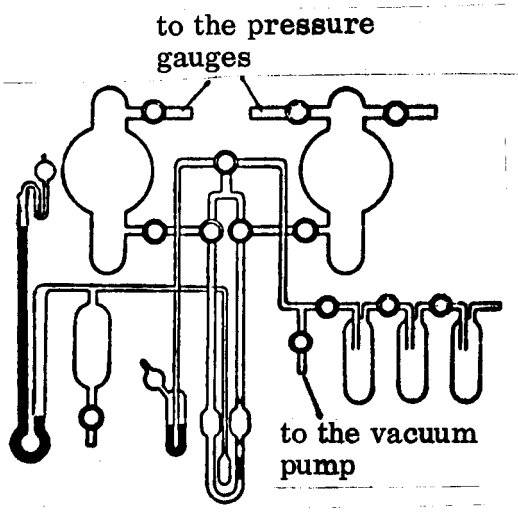

Figure 1.

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The measurements were carried out in three viscosimeters; Table 2 shows the relevant dimensions.

The data shown indicate that the accuracy of the viscosity measurements is about 1.4%. The main source of error lies in the uncertainty in the determination of the diameter of the capillary tube. It therefore seemed desirable to check the viscosimeter-constant with a liquid of accurately known viscosity. The following procedure was used: viscosimeter #1 was calibrated at 20.0° with ethyl ether of a high degree of purity. At this temperature, the viscosity of ethyl ether was

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TABLE 2.

Viscosimeter #	Volume (cm^3)	Diameter of the capillary tube	Length (cm)
1	1.584 ± 0.001	0.00786 ± 0.00002	3.07 ± 0.01
2	1.513	0.00774	3.24
3	1.391	0.00718	3.42

Commas represent decimal points.

taken to be equal to 0.002448 as quoted by Thorpe and Rodger. The resulting viscosimeter constant was assumed to be correct, while the constant that was computed directly from the physical dimensions differed by 0.25%. The viscosity of liquid oxygen was measured very carefully (including the thermal-expansion effect) with this viscosimeter. The average of a large number of observations yielded a value of 0.00190. Then the other viscosimeters were calibrated with liquid oxygen.

Results of the Experiments

Nitrogen was produced by heating a solution of ammonium sulfate with a solution of sodium nitrite. The first batch of gas, about 10 - 15%, was removed through the exhaust. The next batch was condensed in the first trap after impurities had been frozen out in a liquid-air trap. About 50% of the nitrogen was distilled from the first trap to a second one, the temperature of which was slightly below that of the first. The first fraction of the second trap was then condensed into the viscosimeter. Approximately 10 - 12 cm^3 of liquid nitrogen accumulated in the first trap; of this, about 1.5 cm^3 reached the viscosimeter. /474

TABLE 3. NITROGEN

T abs.	η	T abs.	η
77.33	0.00158 CGS	69.1	0.00231 CGS
76.1	0.00165	64.8	0.00284
71.4	0.00209	64.3	0.00290
69.25	0.00228	63.9	0.00292

Commas represent decimal points.

Results of the experiments are given in Table 3 (Fig. 2).

Carbon monoxide was obtained by heating a mixture of sulfuric acid and formic acid. The method of distillation was the same as for nitrogen. Results of the measurements are presented in Table 4 (Fig. 2).

Argon was available in a steel tank (from the Linde Company). It was condensed in a liquid-nitrogen trap, solidified, and the vapor about it was evacuated. The first batch of the remaining argon was transferred into the viscosimeter. The freezing point of argon which was purified by this procedure coincides with the published melting point of pure argon to within a tenth of a degree. Results of the measurements are listed in Table 5 (Fig. 2).

Potassium permanganate was dried well and evacuated; it was used to produce oxygen. The first batch of gas was pumped off, the next was condensed in a trap and distilled. Roughly 50% of the oxygen was then transferred from the first trap into a second one by very slow evaporation. The second trap had a slightly lower temperature; this oxygen was discarded. The oxygen which re- /475

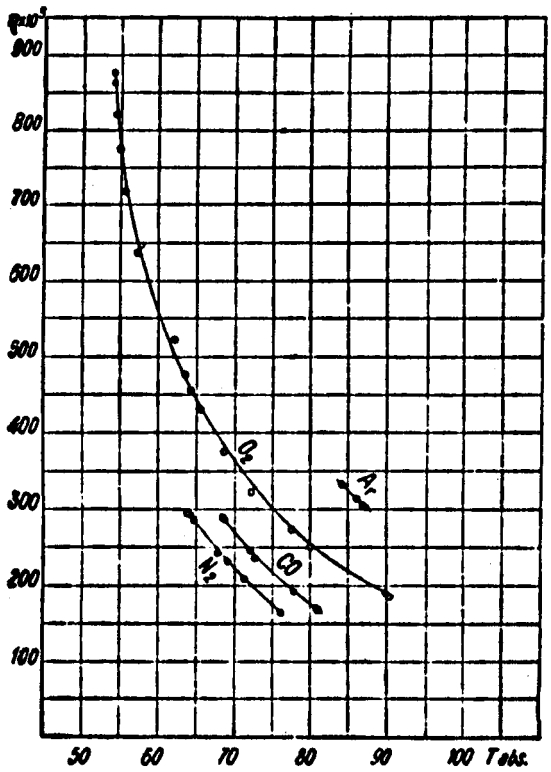

Figure 2.

TABLE 4. CARBON MONOXIDE

T abs.	η	T abs.	η
80.9	0.00170 CGS	72.0	0.00244 CGS
80.8	0.00171	68.6	0.00286
77.7	0.00192	68.55	0.00287
72.4	0.00237	—	—

Commas represent decimal points.

TABLE 5. ARGON

T abs.	η	T abs.	η
87.3	0.00252 CGS	86.25	0.00262 CGS
86.9	0.00256	84.2	0.00280

Commas represent decimal points.

mained in the first trap then reached the carefully evacuated viscosimeter where a well-defined amount condensed.

Wahl [3] found that liquid oxygen stiffens at a temperature just above the triple point. In the course of their determination of the melting point of oxygen, Kamerlingh Onnes and Crommelin [4] have not found this anomaly. It seemed of interest to examine these results by measuring the viscosity at temperatures approaching that of the melting point. The results are listed in Table 6 (Fig. 2).

The measurements show a strong increase of the viscosity with decreasing temperature; however, one definitely cannot say that the liquid becomes stiff since its viscosity is still below that of water. But it is possible that small amounts of impurities have a strong influence on the viscosity of oxygen in the vicinity of the triple point. We have therefore measured the viscosity of very pure oxygen and of oxygen with 2 - 2.5% nitrogen, both at 54.5°K. The viscosity of pure oxygen agreed well with that given in the table, while that of impure oxygen was equal to 0.00829, i.e., differing slightly from that of pure oxygen. Wahl's results can be explained by assuming that liquid oxygen (which can be cooled rapidly with liquid hydrogen) cooled down too much, and actually was below the melting point. According to Wahl, oxygen can be put into a glass-like state by rapid cooling below the melting point. /476

The accuracy of the measurement was reduced slightly at the lowest temperatures because the vapor pressure of oxygen (upon which we based the temperature measurements) is /477

TABLE 6. OXYGEN

T abs.	η	T abs.	P mm	η
90.1	0.00190 CGS	57.4	3.94	0.00648 CGS
80.0	0.00250	57.4	3.62	0.00638
77.4	0.00273	56.4	2.66	0.00717
72.3	0.00323	54.9	1.96	0.00772
68.9	0.00377	54.6	1.76	0.00821
65.4	0.00435	54.5	1.60	0.00863
63.5	0.00476	54.4	1.55	0.00873
61.7	0.00521	—	—	—
59.7	0.00631	—	—	—

Commas represent decimal points.

only known down to about 10 mm Hg [5]. The lowest temperatures were determined by an extrapolation where we assumed the triple point to occur at 54.3°K [6]. In Table 6 we list the oxygen vapor pressures from which the temperatures were determined.

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